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## Chemical contaminants in the Wadden Sea: Sources, transport, fate and effects



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#### ABSTRACT

The Wadden Sea receives contaminants from various sources and via various transport routes. The contaminants described in this overview are various metals (Cd, Cu, Hg, Pb and Zn) and various organic contaminants (polychlorinated biphenyls (PCBs), polycyclic aromatic hydrocarbons (PAHs) and lindane (hexachlorocyclohexane,  $\gamma$ -HCH)). In addition, information is presented about other and emerging contaminants such as antifouling biocides (e.g. TBT and Irgarol), brominated flame retardants (BFRs), poly- and perfluorinated compounds (PFCs) and pharmaceutical and personal care products (PPCPs).

Special attention is given to biogeochemical processes that contribute to the mobilization of contaminants in the surface sediments of the Wadden Sea. Finally, the effects on organisms of contaminants are reviewed and discussed.

The main source of contaminants in the Wadden Sea are the rivers Rhine (via de Dutch coastal zone), Elbe and Weser. The Wadden Sea is not a sink for contaminants and adsorbed contaminants are transported from east to west. The surface sediments of the Wadden Sea are an important source for contaminants to the water above. The input and concentration of most contaminants have significantly decreased in water, sediments, organisms (e.g., mussel, flounder and bird eggs) in various parts of the Wadden Sea in the last three decades. Remarkably, the Cd concentration in mussels is increasing the last decades.

In recent decades, the effects of contaminants on organisms (e.g., flounder, seal) have fallen markedly. Most of the affected populations have recovered, except for TBT induced effects in snails. Little is known about the concentration and effects of most emerging contaminants and the complex environmental mixtures of contaminants. It is recommended to install an international coordinated monitoring programme for contaminants and their effects in the whole Wadden Sea and to identify the chemical contaminants that really cause the effect.

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#### 1. Introduction

Coastal seas such as the Wadden Sea are often described as an ultimate and permanent sink for contaminants in the sediments (Gerlach, 1976; Lozán et al., 2002; Wolff, 1988). The idea of a sink is that once the contaminants have been taken up by the sediments they are immobilized and so are no longer available for biological processes. Studies on chemical time bombs (e.g., Flyhammar and Håkansson, 1999; Salomons and Stigliani, 1995; Stigliani, 1991) and on the behaviour of contaminants in dumped dredging spoil (e.g., Förstner et al., 1984; Salomons and Förstner, 1988), however, have shown that there is no guarantee that sediments are a secure repository for contaminants. These authors demonstrated that changes in external conditions (e.g., climate) and in the internal physical and biogeochemical conditions in the sediments and dredging spoil (e.g., pH, salinity and redox potential) could result in particles and dissolved

contaminants being mobilized and entering the overlying water (Chester, 1993; Förstner et al., 1984; Pakhomova et al., 2007; Stigliani, 1988; Vink et al., 2010a,b; Williams et al., 1994).

In this paper, a review is presented on the sources, transport, fate and effects of contaminants in the Wadden Sea. The response of the Wadden Sea system to the input of contaminants and the contribution of the most important physical and biogeochemical processes to the ultimate fate of these contaminants in the Wadden Sea will be discussed using the available data and information.

#### 2. The Wadden Sea

The Wadden Sea is a shallow sea with tidal flats off The Netherlands, Germany and Denmark (Fig. 1). It is a semi-enclosed water body, with connections with the coastal zone of the North Sea between its fourteen barrier islands (Fig. 1). From the early work of De Groot (1963, 1966, 1973), Duinker et al. (1974), Vandeginste and Salemink (1976) and Förstner (1979) it is known that due to the open connection of the Wadden Sea with the North Sea, seawater and Wadden seawater

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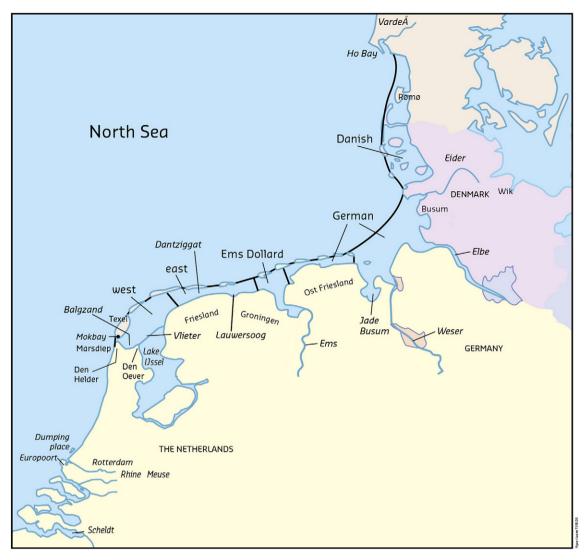


Fig. 1. Map of the Dutch coastal zone and the Wadden Sea. The Dutch part of the Wadden Sea is divided into three areas: West, East and Ems-Dollard.

containing dissolved and particulate contaminants are exchanged during every tidal cycle.

The Wadden Sea is less saline than the North Sea because its seawater is diluted by freshwater originating mainly from the rivers Rhine, Ems, Weser, Elbe, Eider and Varde Å, and Lake IJssel. The German rivers (Ems, Elbe and Weser), Danish rivers (e.g., Varde Å) and water from Lake IJssel discharge directly into the Wadden Sea via their estuaries or sluices. The dissolved and particulate contaminants of the Rhine and Meuse rivers enter the western Wadden Sea via the Dutch coastal zone and Lake IJssel (Postma, 1980, 1981; Ridderinkhof, 1990; Verlaan and Spanhoff, 2000). Zimmerman and Rommets (1974) calculated that in the western part of the Wadden Sea 7% of the water is Rhine water and 15% of the water is from Lake IJssel. The average residence time of fresh water within the western Wadden Sea is 13 tidal periods (Postma, 1954; Ridderinkhof, 1990).

In general, suspended particulate matter (SPM) in the Wadden Sea originates from the rivers and sluice systems and from local primary production (Eisma, 1993; Salomons and Eysink, 1983). Eisma (1983) estimated that the average residence time of SPM in the water of the western Wadden Sea was about 43 days. Postma (1954, 1961) and as long ago as the mid-20th century Van Straaten and Kuenen (1957, 1958) showed that the Wadden Sea accumulates SPM from the surrounding North Sea and rivers. They estimated that about 50% of the SPM from the Rhine is trapped in the Wadden Sea (Postma, 1961). Various studies have presented annual amounts of SPM entering the

western Wadden Sea. The net annual amount of SPM entering the Wadden Sea via the Marsdiep tidal inlet in the western Wadden Sea varies between 0.8 and 3.6 million m³ per year (see Eisma, 1966, 1982, 1993; Salomons and Eysink, 1983; Van Duren and Van der Valk, 2010; Verhagen, 1990 and references cited). However, from the ferry box measurements in the Marsdiep it is known that the annual amount is much larger than the earlier estimates: it is around 5–10 Mt per year (Tiessen et al., 2012). These larger amounts are in the same order of magnitude as the estimates made by Brolsma (1982) and Eysink (1979) of loads and fractions of silt in the surface sediments.

The SPM originating from the Rhine and Meuse is concentrated in a narrow strip along the Dutch coast due to estuarine circulation processes (Postma, 1981; Suijlen and Duin, 2001; Verlaan and Spanhoff, 2000). Some of the SPM and associated contaminants in the Dutch coastal zone originates from the dumping of dredged harbour spoil in the Dutch coastal zone (Salomons and Förstner, 1988; Yland et al., 2000). De Kok et al. (1992) showed that the dumping of dredge spoil from Rotterdam harbours in the Dutch coastal zone is an important source of SPM and the associated contaminants in the western Wadden Sea. The annual amount of dredge spoil in the Dutch coastal zone varies: it was only  $4*10^6$  m³ in 1962 but reached around  $25*10^6$  m³ in 1968 and in 1983 (De Kok et al., 1992). About 33% of the dumped spoil finds its way back to Rotterdam harbours by prevailing currents, 20% remains at the dumping site (Louisse, 1986; Louisse, 1987) and the rest is transported north to the Wadden Sea (De Kok et al., 1992). There is a

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